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Washington, D.C. 20231



Signature of Person Undertaking Mailing

Docket No.: 2734/MEINKE/US

Date: August 12, 1999

Sir:

Transmitted herewith for filing is the patent application of Dr. Michael Benje

(Name(s) of Inventor(s))

**FOR:** Process for Removing Fine Dust from a Fluidized-bed Reactor, in particular for the Oxychlorination of Ethylene

(Title of Application)

ENCLOSED ARE:

- (X) Specification ( 7 pages), Claims ( 2 pages / 9 claims\*) & Abstract: Yes X 2 Sheets of Drawing(s);  
(X) Declaration and Power of Attorney EXECUTED? Yes X No       
(X) Assignment to Krupp Uhde GmbH,  
Friedrich-Uhde-Straße 15, 44141 Dortmund, Germany  
(X) Certified copy of Application No. 198 37 957.9

the priority of which is claimed under 35 USC 119;

Verified Statement to establish Small Entity Status under 37 CFR 1.9 and 1.27

Information Disclosure Statement, PTO-1449 and                      references;

Preliminary Amendment

\*Please calculate fees after taking into account the Preliminary Amendment

**THE FILING FEE HAS BEEN CALCULATED AS SHOWN BELOW:**

	Claims filed	Extra	SMALL ENTITY or LARGE ENTITY	
Basic Fee			\$ 380.00	\$ 760.00
Total Claims	<u>12</u> - 20 =	X \$9.=	X \$18. =	
Indep. Claims	<u>2</u> - 3 =	X \$39.=	X \$78.=	
Multiple Dep. Claim Presented?		X \$130.=	X \$260.=	
Total Filing Fee:			\$	\$ 760.00
Assignment recordal fee (\$40.00):			\$	\$ 40.00
PLEASE CHARGE:			\$	\$ 800.00

( ) Enclosed is a check for the fees indicated above.

The Commissioner is hereby authorized to charge payment of the following fees associated with this communication and credit and overpayment to Deposit Account No. **06-0923**. A duplicate copy of this sheet is enclosed.

- (X) The fees indicated above.  
(X) Any additional filing fees required under 37 CFR 1.16.  
(X) Any filing fees under 37 CFR 1.16 for the presentation of extra claims.

The Commissioner is hereby authorized to charge payment of the following fees during the pendency of the application or credit any overpayment to Deposit Account No. **06-0923**. A duplicate copy of this sheet is enclosed.

- (X) Any additional filing fees required under 37 CFR 1.17.  
(X) Any filing fees under 37 CFR 1.16 for the presentation of extra claims.

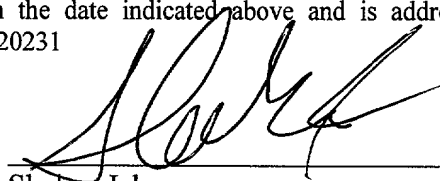
Respectfully submitted  
for Applicant.Shahan Islam  
Reg. No. 32,507

Friedman Siegelbaum LLP  
Seven Becker Farm Road  
Roseland, NJ 07068-1757

ST. 8/23/99  
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Shahar Islam

2734/Meinke

-----X	
Applicant:	Michael Benje
Serial:	Not Assigned
Filed:	August 22, 1999
For:	PROCESS FOR REMOVING FINE DUST FROM A FLUIDIZED-BED REACTOR, IN PARTICULAR FOR THE OXYCHLORINATION OF ETHYLENE
-----X	

Examiner: Not Assigned  
Group Art Unit: Not Assigned

Honorable Commissioner of  
Patents and Trademarks  
Washington, D.C. 20231

**PRELIMINARY AMENDMENT**

Prior to an action on the merits, please amend the above-identified application as follows:

**IN THE CLAIM:**

Please amend claims 1-9 and add claims 10-12 as follows:

**Patent claims:**

1. (Amended) A process for removing fine dust from a fluidized-bed reactor, [in particular for the oxychlorination of ethylene], said process comprising the steps of: [wherein]

removing the fine dust inside [the] a reactor [is removed] via filter cartridges[, in particular sintered metal filter cartridges,]; and

passing [the] reaction gas mixture [is passed] to [the] a quench from a dome of the reactor [dome].

2. (Amended) The process as claimed in claim 1, further comprising the step of [wherein] removing a part-stream in the form of a bypass stream having a predetermined fine dust fraction below a predetermined particle size [is removed] from the reactor in addition to the main stream.

3. (Amended) The process as claimed in claim 1 [or 2], wherein the main stream and the bypass stream are removed from separate dome spaces of the reactor.

4. (Amended) The process as claimed in claim 1, further comprising the steps of: [or in any of the following claims, wherein the bypass stream is switched on and off according to an analysis of]

analyzing a catalyst sample and/or a change in the heat transfer and/or a deterioration of the fluidization behavior[.]; and

switching on or off the bypass stream according to said analysis.

5. (Amended) A fluidized-bed reactor[, in particular for the oxychlorination of ethylene, with the use of] using catalyst granules subjected to abrasion, said reactor comprising: [wherein]

at least one baseplate [(4)] having [sintered metal] filter cartridges [(5) is provided] in a [the] dome [(6)] of the reactor (1), wherein the filter cartridges are dipppable [optionally dipping] into [the] an upper region of the fluidized bed [(2)].

6. (Amended) The fluidized-bed reactor as claimed in claim 5, wherein the dome space [(6)] is divided, above the plate carrying the filter cartridges [(5)] on its lower surface, into at least two chambers [(6, 6a)], each having an outlet [(11)] for a main stream to the quench and a bypass stream [(8)].

7. (Amended) The fluidized-bed reactor as claimed in claim 6, wherein the filter elements [(5a)] coordinated with the bypass [(8)] have pore size differing from that of the fine dust filter cartridges [(5),] for the controlled passage of fine dust fractions.

8. (Amended) The fluidized-bed reactor as claimed in claim 5[ or in any of the following claims], wherein the ratio of filter elements [(5a)] allowing through fine dust to filter cartridges [(5)] retaining the fine dust is [in the region of] approximately 1:9.

9. (Amended) The fluidized-bed reactor as claimed in claim 5 [or in any of the following claims] [wherein the baseplate (4) is provided with] further comprising a cleaning means using [by means of] compressed gas pulses on the baseplate.

-- 10. (New) The process of claim 1, wherein said process is used primarily for the oxychlorination of ethylene.

11. (New) The process of claim 10, wherein said cartridges are sintered metal filter cartridges.

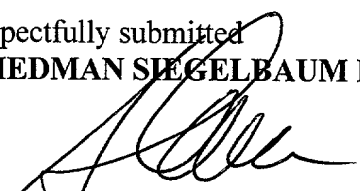
12. (New) The fluidized-bed reactor of claim 5, wherein said filter cartridges are sintered metal filter cartridges.- -

**REMARKS**

The purpose of the Preliminary Amendment was to remove some of the improper multiple dependencies from the claims and to conform them better to U.S. practice.

The amendment does not constitute a relinquishment of any subject matter; applicant reserves the right to bring back any of the same dependencies existing prior to this amendment.

Respectfully submitted  
**FRIEDMAN SIEGELBAUM LLP**

  
Shahan Islam (Reg. No. 32,507)

8/23/99 23  
Dated: August 23, 1999

**Friedman Siegelbaum LLP**  
Seven Becker Farm Road  
Roseland, New Jersey 07068  
(973) 992-1990, Ext. 191

*Application for Letters Patent  
of the United States*

*Inventor:*

DR. MICHAEL BENJE

*Title of Invention:*

PROCESS FOR REMOVING FINE DUST FROM A FLUIDIZED-BED  
REACTOR, IN PARTICULAR FOR THE OXYCHLORINATION  
OF ETHYLENE

*To All Whom It May Concern:*

*The following is a specification  
of the aforesaid Invention.*

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PROCESS FOR REMOVING FINE DUST FROM A FLUIDIZED-BED REACTOR,  
IN PARTICULAR FOR THE OXYCHLORINATION OF ETHYLENE

The invention relates to a process for removing fine dust from a fluidized-bed reactor, in particular for the oxychlorination of ethylene.

There is a number of known processes in which chemical reactions are initiated by means of catalysts in a fluidized bed, for example in oxychlorination, in which ethylene-oxygen and HCl are reacted in a fluidized-bed reactor over a copper-containing catalyst to give 1,2-dichloroethane and water.

In such fluidized-bed processes, abrasion of the fluidized-bed particles inevitably occurs, resulting in fine dust in the fluidized bed. Since these fine dust particles are entrained by the reaction gas mixture, they can be separated off inside or outside the reactor, for example by cyclones connected in series or by fine dust filters, for example by hoses of a Gore-Tex membrane on PTFE needle felt.

German Laid-open Specification 20 19 210 and also DE-40 30 086-C1 disclose fluidized-bed reactors and fluidized-bed processes, respectively, comprising filter cartridges, arranged inside the reactor, for the exit gas. Such filter cartridges invariably allow small amounts of fine dust to pass through (cf. expert opinion DE-21 66 912-A1, in particular the last paragraph of the description of this publication).

Owing to the physical circumstances of the

fluidized beds used, cyclone inlets must be arranged at a certain minimum height above the fluidized bed since from this height onward the dust content in the gas taken off is approximately constant, and about 25% of the height of the reactor may be stated as an example without the invention being restricted to these dimensions.

It is known that efforts have been made to replace the cyclones and the downstream fine dust filters with a single fine dust filter arranged directly in the reactor, it being possible to clean the filter cartridges used by means of compressed gas pulses from the series side. The filters which are used may dip directly into the fluidized bed, which can inevitably lead to a reduction in the height of reactors.

Furthermore, such a procedure should permit complete dust separation.

A particular disadvantage of this procedure is, however, that fine dust fractions which inevitably differ considerably from the desired spherical shape of the fluidized-bed particles inevitably accumulate in the fluidized bed, so that the characteristics of the fluidized bed change. While a very small amount is useful for better fluidization of the fluidized bed, larger amounts can lead to a transition from an effervescent fluidized bed to a percussive fluidized bed or to coating of the cooling surfaces, which leads to a gradual deterioration and finally

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to the collapse of heat transfer.

It is here that the present invention is of use, its object being to permit specific control of the amount of fine dust particles in a reactor while reducing the height of the reactor and at the same time filtering off the fine dust from the main stream leading to the quench.

This object according to the invention is achieved by a process of the type defined at the outset if the fine dust inside the reactor is removed via filter cartridges, in particular sintered metal filter cartridges, having a defined larger pore size than those of the fine dust filter, and the reaction gas mixture is passed to the quench from the reactor dome, it being possible in a further embodiment to ensure that a part-stream in the form of a bypass stream having a predetermined fine dust fraction below a predetermined particle size is removed from the reactor in addition to the main stream.

By means of the procedure according to the invention, it is possible to establish exactly the desired fine dust fraction in the fluidized-bed reactor. If, for example by sieve analysis of a catalyst sample, it is found that the fine dust fraction in the reactor increases above a predeterminable, permissible level, the bypass line and the coordinated filter cartridges can be opened and fine dust fractions below a specific minimum particle size thus

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gradually leave the reactor.

In a further embodiment, it is envisaged that the main stream and the bypass stream are removed from separate dome spaces of the reactor, it being possible, instead of separate dome spaces, also to provide groups of filter baseplates having filter cartridges, which are then controlled in different ways.

The bypass stream can be switched on and off on the basis of different criteria, for example on the basis of a corresponding, already mentioned analysis of a catalyst sample, on the basis of the change in the heat transfer or the deterioration of the fluidization behavior of the fluidized bed, for example if an abnormal high density of the fluidized bed is found.

To achieve the object set, the invention also provides a fluidized-bed reactor, in particular for the oxychlorination of ethylene, with the use of catalyst granules subjected to abrasion, e.g.  $\gamma$ -alumina having a mean particle size of from about 40 to 60  $\mu\text{m}$ , which is distinguished by the fact that at least one baseplate having filter cartridges, in particular sintered metal filter cartridges, is provided in the dome of the reactor, it being possible for the filter cartridges to dip into the upper region of the fluidized bed.

At this point, it may be noted that the filter

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cartridges may consist of another material, e.g. ceramic, and in this respect the invention is not restricted to said sintered metal filter cartridges.

An embodiment provides a fluidized-bed reactor which is distinguished by the fact that the dome space is divided, above the plate carrying the filter cartridges on its lower surface, into at least two chambers, each having an outlet for a main stream to the quench and a bypass stream.

The division of the plate carrying the filter cartridges into two regions which are separated from one another in the dome space has the advantage that the bypass stream can be switched on and off separately without greatly influencing the other filters.

Instead of such a division, the filter cartridges may also be grouped together in bundles, combined and inserted from the outside into the dome space and flange-mounted there. Such a bundle can then be switched, for example, as a bypass.

The invention also envisages that the filter elements coordinated with the bypass have a pore size differing from that of the fine dust filter cartridges, for the controlled passage of fine dust fractions.

The ratio of filter elements allowing through fine dust to filter elements retaining fine dust may be in the region of 1:9, but the invention is not restricted to these

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numbers and other ratios may also be provided here.

The plate or plates carrying the filter cartridges can be provided with a cleaning means by means of compressed gas pulses.

The invention is illustrated in more detail below with reference to the drawing by way of example.

Fig. 1 and 2 of the drawing show greatly simplified sections of reactor dome spaces with variants of the coordination, according to the invention, of filter cartridges.

A reactor, reproduced only in simplified form in the Figure and denoted generally by 1, has a fluidized bed 2 which is indicated in Fig. 1 only by its upper limit and in which, for example,  $\gamma$ -alumina particles having a mean particle size of from 40 to 60  $\mu\text{m}$  are present, in order to react ethylene-oxygen and HCl to give 1,2-dichloroethane and water.

In the example of Fig. 1, the fluidized-bed reactor 1 has, in its dome denoted by 3, a dome plate 4 which carries a multiplicity of filter cartridges 5 which point toward the fluidized bed 2 and, depending on the design, can optionally also dip into the fluidized bed 2.

In the example of Fig. 1, the dome space denoted by 6 is separated by a partition 7 into a larger region and a smaller region 6a, a small number of filter cartridges 5a

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A corresponding gas stream comprising fine dust particles is fed via a bypass line 8, for example to an external fine dust filter 9, and the fine dust can then be discharged and disposed of, which is indicated by a container 10, and the gas can then be fed via the bypass line 8 back to the main stream, denoted by 11.

Here, too, the filter cartridges 5a may once again have a pore size sufficiently large for fine dust to be removed from the dome space 6 via the bypass line 8; here, as in the embodiment of Fig. 1, separation is effected via an external fine dust filter 9.

Of course, the embodiments of the invention which are described can also be modified in various respects without departing from the basic concepts; for example, a combination of the designs according to Fig. 1 and 2 can be implemented, etc.

Patent claims:

1. A process for removing fine dust from a fluidized-bed reactor, in particular for the oxychlorination of ethylene, wherein the fine dust inside the reactor is removed via filter cartridges, in particular sintered metal filter cartridges, and the reaction gas mixture is passed to the quench from the reactor dome.
2. The process as claimed in claim 1, wherein a part-stream in the form of a bypass stream having a predetermined fine dust fraction below a predetermined particle size is removed from the reactor in addition to the main stream.
3. The process as claimed in claim 1 or 2, wherein the main stream and the bypass stream are removed from separate dome spaces of the reactor.
4. The process as claimed in claim 1 or in any of the following claims, wherein the bypass stream is switched on and off according to an analysis of a catalyst sample and/or a change in the heat transfer and/or a deterioration of the fluidization behavior.
5. A fluidized-bed reactor, in particular for the oxychlorination of ethylene, with the use of catalyst granules subjected to abrasion, wherein at least one baseplate (4) having sintered metal filter cartridges (5) is provided in the dome (6) of the reactor (1), the filter

cartridges optionally dipping into the upper region of the fluidized bed (2).

6. The fluidized-bed reactor as claimed in claim 5, wherein the dome space (6) is divided, above the plate carrying the filter cartridges (5) on its lower surface, into at least two chambers (6, 6a), each having an outlet (11) for a main stream to the quench and a bypass stream (8).

7. The fluidized-bed reactor as claimed in claim 6, wherein the filter elements (5a) coordinated with the bypass (8) have a pore size differing from that of the fine dust filter cartridges (5), for the controlled passage of fine dust fractions.

8. The fluidized-bed reactor as claimed in claim 5 or in any of the following claims, wherein the ratio of filter elements (5a) allowing through fine dust to filter cartridges (5) retaining the fine dust is in the region of 1:9.

9. The fluidized-bed reactor as claimed in claim 5 or in any of the following claims, wherein the baseplate (4) is provided with a cleaning means by means of compressed gas pulses.

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[illegible]

This is achieved if the fine dust inside the reactor is removed via filter cartridges, in particular sintered metal filter cartridges, and the reaction gas mixture is passed to the quench from the reactor dome, a part-stream in the form of a bypass stream having a predetermined fine dust fraction below a predetermined particle size being removed from the reactor in addition to the main stream.

Drawing to be published herewith: Fig. 1.



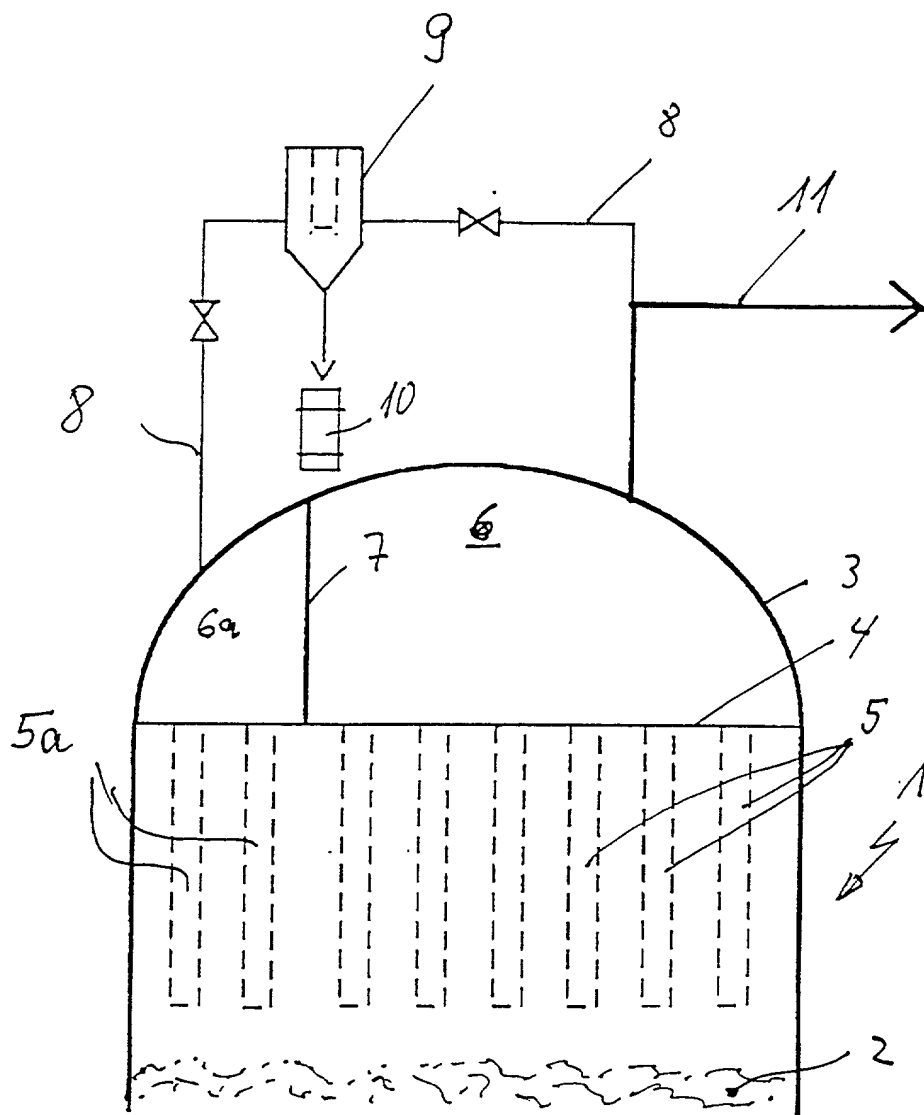


Fig. 1

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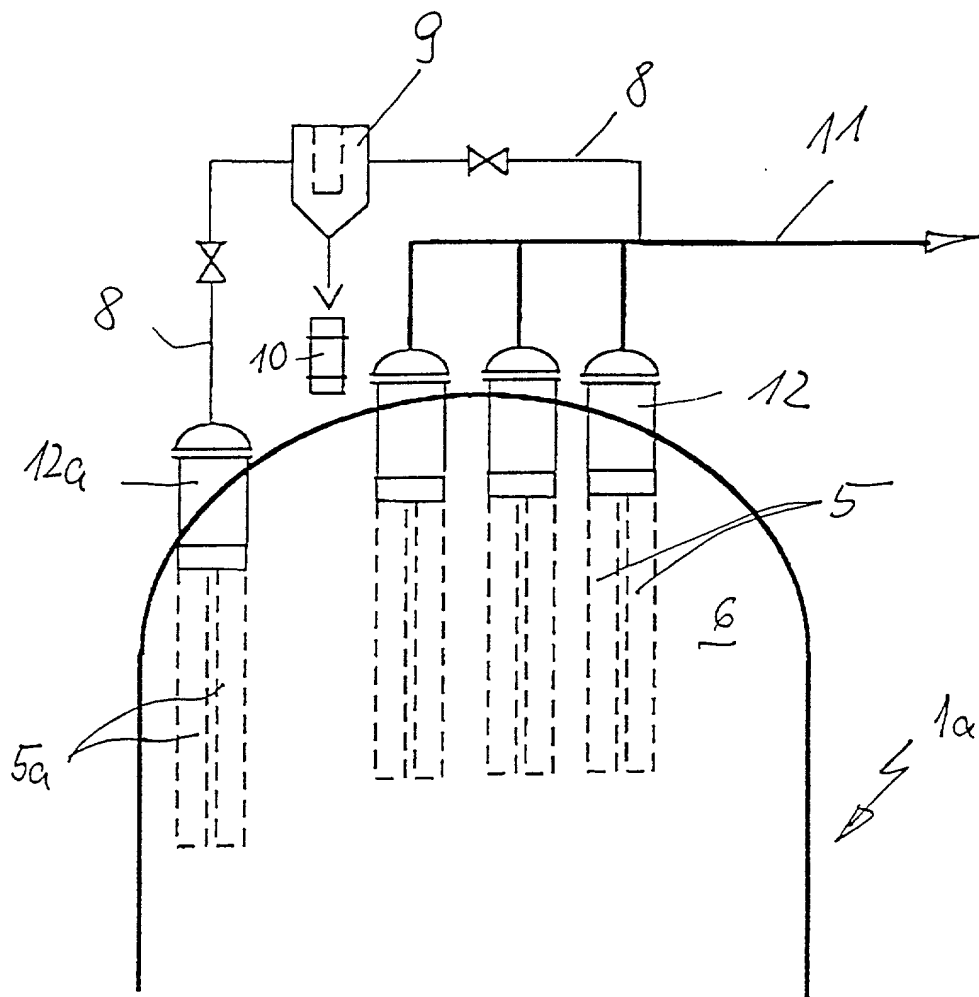


Fig. 2

**FRIEDMAN □ SIEGELBAUM**  
**DECLARATION AND POWER OF ATTORNEY**

I, a below named inventor, I hereby declare that:

My residence, post office address and citizenship are as stated below my name.

I believe I am the original, first and sole inventor (if only one name is listed below), or an original, first and joint inventor (if more than one inventor's name is listed below), of the subject matter which is claimed and for which a patent is sought on the invention entitled Process for Removing Fine Dust from a Fluidized-bed Reactor, in particular for the Oxychlorination of Ethylene

Title of Invention

The specification of which: ☒ is attached hereto ☐ was filed on \_\_\_\_\_ Application Serial No. \_\_\_\_\_  
(for declaration not accompanying application)

With amendment(s) filed on \_\_\_\_\_  
(date(s) of all amendment(s))

I hereby state that I have reviewed and understand the contents of the above identified specification, including the claims, as amended by any amendment referred to above.

I acknowledge the duty to disclose information which is material to the examination of this application in accordance with Title 37, Code of Federal Regulations, §1.56(a).

I hereby claim foreign priority benefits under Title 35, United States Code, §119/§172 of any foreign application(s) for patent or inventor's certificate listed below and have also identified below any foreign application for patent or inventor's certificate having a filing date before the date of the application on which priority is claimed:

**EARLIEST FOREIGN APPLICATION(S), IF ANY, FILED PRIOR TO THE FILING DATE OF THE APPLICATION**

APPLICATION NUMBER	COUNTRY	DATE OF FILING (Day, Month, Year)	PRIORITY CLAIMED UNDER 35 U.S.C. 119/172
198 37 957,9	Germany	August 21, 1998	Yes <input checked="" type="checkbox"/> No <input type="checkbox"/>
			Yes <input type="checkbox"/> No <input type="checkbox"/>
			Yes <input type="checkbox"/> No <input type="checkbox"/>

I hereby claim the benefit under Title 35, United States application(s) listed below and, insofar as the subject matter of each of the claims of this application is not disclosed in the prior United States application in the manner provided by the first paragraph of Title 35, United States Code §112, I acknowledge the duty to disclose material information as defined in Title 37, Code of Federal Regulations, §1.56(a) which occurred between the filing date of the prior application and the national or PCT international filing date of this application:

APPLICATION NUMBER	FILING DATE	STATUS		
		PATENTED	PENDING	ABANDONED

**POWER OF ATTORNEY:** As a named inventor, I hereby appoint Stanley I. Rosen (Reg. No. 19,441), Richard I. Samuel (Reg. No. 34,435), Shaban Islam (Reg. No. 32,507), Theresa A. O'Rourke (Reg. No. 40,747) and Hugh H. Jacobson (Reg. No. 44,799) whose address

mad Siegelbaum, Seven Becker Farm Road, Roseland, New Jersey, 07068-1757 as my attorneys, to prosecute this application, respect all business in the U. S. Patent and Trademark Office connected therewith.

Correspondence To: SWAILAN ISLAM, ESQ. Direct Telephone Number (973) 992-1990, EXT. 191  
 FRIEDMAN SIEGELBAUM, Seven Becker Farm Road, Roseland, New Jersey 07068-1757

Name of Inventor	Last Name Benje	First Name Michael	Middle Name
Place of Birth	City Darmstadt	State of Foreign Country Germany	Country of Citizenship Germany
Address	No. and Street Address Berkstrasse 6	City Darmstadt	State or Country Germany
			Zip Code 64280

I declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 10 of the United States Code and that such willful false statements may invalidate the validity of this application or any patent issuing thereon.

Signature of Inventor	<i>Michael Benje</i>		
Date	16.08.99		